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NUCLEATION IN METASTABLE SOLID SOLUTION – STOCHASTIC KINETIC MEAN FIELD APPROACH VERSUS CLASSICAL NUCLEATION THEORY*

The application of Stochastic Kinetic Mean Field approach to the kinetics of nucleation at the decomposition of supersaturated binary solid solution is presented. The dependencies of incubation time on the noise amplitude and the supersaturation are obtained. SKMF modeling demonstrates the validity of Classical Nucleation Theory. The logarithm of nucleation time is inversely proportional to the squared supersaturation. The logarithm of nucleation time is a linear function of the inverse squared noise amplitude.

Keywords: SKMF approach, mean field, binary solid solution, decomposition, nucleation, incubation time.

1. Introduction

Nucleation stage of the first-order phase transformations in alloys is crucial for prediction of mechanical, electrical, magnetic properties of multiphase materials. During last decades the new experimental possibilities have been developed enabling direct observation of nuclei formation during aging and solid-state reactions – for example, 3D TAP, HR TEM, AFM [1–3]. Yet many details of the nucleation stage still remain the mystery. For example, nucleation of the new phase means the change of composition, size and (often) structure. It seems strange but in many cases, we still do not know the sequence in changing these properties. In general, Classical Nucleation Theory (CNT) takes for granted that the nuclei at once appears with optimal composition and structure and then changes only the size during precipitation. On the other hand, Schmelzer and Abyzov demonstrated by numerical solving of Master Equations that initially the undercritical embryo may change the size and only later it changes the composition [4, 5]. In the case of intermetallic compound nucleation, the situation is more interesting because, besides composition change, the

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nucleation includes also ordering. As far as we are informed, the sequence of ordering and composition changes in most cases is still unknown. Two most widespread methods of the nucleation kinetics investigation are Fokker-Plank approach and Monte Carlo (MC) simulation [6]. Fokker-Plank approach seems a good solution but it contains a number of not very well determined parameters (attachment and detachment frequencies) and not very well proved phenomenological assumptions (for example, steady state diffusion around precipitate). Monte Carlo is more direct and atomistic, but the level of fluctuations in this method is so high that it is very difficult to distinguish the structures in small volumes. So, we need some transient method between atomistic MC and mean-field phenomenological approach. Recently, our group, jointly with the group of Debrecen University, developed the new method called Stochastic Kinetic Mean Field (SKMF) [7–9]. This method combines George Martin's mean field atomistic approach with the noise of local atomic fluxes. This method is not far from Khachatryan's approach [10]. Khachatryan's approach is based on linear thermodynamics and fluctuation-dissipation theorem introducing the noise of concentration and local order. SKMF approach is inherently nonlinear and therefore applicable to the early stages of solid-state reactions under sharp concentration gradient. In this approach, the probability of atomic exchanges is proportional to the difference of exponents of chemical potentials, instead of a difference of just chemical potentials. Moreover, noise is introduced directly into atomic jumps quantity, instead of the noise of composition. Also, no special order noise is introduced. The fluctuation of local order at any site is determined by the change of atomic probabilities at each site at the atomic scale. Recently we proved rigorously [11] that without noise Martin's equations and their 3D modification [12] may lead only to decrease of the free energy. For overcoming the nucleation barrier one may introduce the noise of frequency with some amplitude A [7]. For the particular case of the ideal solid solution, we proved that the introduction of the noise of amplitude A is equivalent to averaging over finite number M copies of the canonical ensemble with

$$M = \frac{1}{c(1-c)A^2}, \quad (1)$$

where c is the average composition of the solution.

In [11] we proved that equation (1) can be also applied to the regular solid solution. In [7] we only indicated the possibility of nucleation modeling with SKMF and presented movie of such process. Here we analyze the nucleation in the metastable solid solution on a more regular basis. We limit ourselves only to nucleation of new solid solutions in parent solid solution. Nucleation of the ordered phases will be discussed elsewhere. The plan of this paper is the following. In section 2 we resemble the algorithm the SKMF model and formulate the criteria of successful nucleation. In section 3 we present the main result of nucleation modeling of the SKMF method. First of all, we determine the dependence of incubation time on the inverse squared noise amplitude and on supersaturation. In section 4 we suggest the application of CNT to nucleation kinetics with an account of composition dependence of the surface energy between parent and nucleating phases. We compare the results with the SKMF approach.

2. Basics of SKMF

We investigate the diffusion-controlled processes at rigid lattice with face-centered cubic (FCC) structure. Technically, FCC structure is realized as a sublattice of the simple cubic lattice with lattice constant $a/2$ with the odd sum of indexes $i+j+k$ along axes x, y, z . To simplify the notations, everywhere below we use the symbol i as the abbreviation of 3 indexes and symbol in as the abbreviation of one of $z=12$ nearest neighboring sites. Each site of this lattice is characterized by so-called "concentration" c_i which is, in fact, the probability of finding species A at this site ($(1-c_i)$ is the probability of finding species B). Mean field approximation means that this probability explicitly does not depend on the neighborhood (but implicitly, it depends). The main equations for the array of concentrations, in case of direct exchange mechanism, are

$$\frac{dc_i}{dt} = -\sum_{in=1}^z c_i (1-c_{in}) (\Gamma_{i \rightarrow in} + \delta \Gamma_{i \rightarrow in}^{Lang}) + \sum_{in=1}^z c_{in} (1-c_i) (\Gamma_{in \rightarrow i} + \delta \Gamma_{in \rightarrow i}^{Lang}). \quad (2)$$

Here the exchange frequencies $\Gamma_{i \rightarrow in}$ and $\Gamma_{in \rightarrow i}$ are determined by the Boltzmann-like exponential expression containing energy before the jump, which self-consistently depends on the concentrations in the neighborhood of both sites making the problem substantially non-linear:

$$\Gamma_{i \rightarrow in} = \Gamma_0 \exp\left(-\frac{E_{i,in}}{kT}\right), \quad (3)$$

$$E_{i,j} = (M - V) \sum_{in=1}^z c_{in} + (M + V) \sum_{jn=1}^z c_{jn}, \quad (4)$$

where $M = (V_{AA} - V_{BB})/2$, $V = V_{AB} - (V_{AA} - V_{BB})/2$ are diffusion asymmetry and mixing energy, V_{AA} , V_{BB} , V_{AB} – pair interaction energies for nearest neighbors. The noise of atomic jumps is realized here as a noise of jump frequencies $\delta \Gamma_{i \rightarrow in}^{Lang}$. Noise distribution can be chosen in various forms, including Gaussian. For simplicity we choose stepwise distribution within interval $(-A\sqrt{3}/\sqrt{dt}, A\sqrt{3}/\sqrt{dt})$ providing the dispersion $\langle (\delta \Gamma_{i \rightarrow in}^{Lang})^2 \rangle = A^2/dt$:

$$\delta \Gamma_{i \rightarrow in}^{Lang} = \frac{A\sqrt{3}}{\sqrt{dt}} (2 \cdot \text{random} - 1). \quad (5)$$

The system of equations (2) was numerically solved for the cubic sample $30 \times 30 \times 30$ sites containing 13500 atoms.

3. Results of SKMF Modeling

In [7] we checked that the phase diagram for the binary system in the KMF approach coincides with the regular solid solution model. The binodal and spinodal for reduced temperature kT/V are presented at figure 1.

For nucleation study, we took initially homogeneous solid solutions with compositions and temperatures in the metastable region (between binodal and spinodal). For each set of parameters (reduced temperature, composition, noise amplitude) we executed 1000 runs, each time waiting of nucleation of the first viable nucleus and then making an average of waiting times over all runs. Apparently, waiting time depends on the system size (the larger the size – the smaller should be waiting time for the first successful nucleation). Therefore, for correct comparison, we made all simulations for the same size (the size effect on nucleation and possible suppression of nucleation by the small size was discussed, for example in [13–15]). We checked several different criteria of nucleation and chose the following one: we calculate the average concentration over each cluster containing 13 sites (1 central and 12 nearest neighbors). We checked that in case of reaching the concentration in one of the sites some value the decomposition becomes inevitable and irreversible. So, in our simulations, we stop calculation of any run just after reaching the average concentration of value 0,5 at any site of the system.

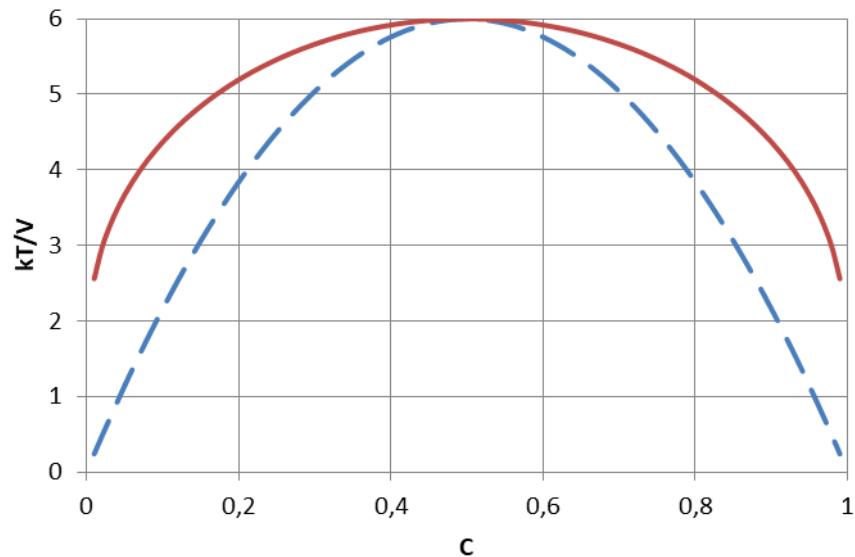


Fig. 1. Phase diagram of a binary solid solution calculated with regular solid solution model. The binodal and spinodal are shown with solid and dashed lines accordingly.

3.1. Dependence of incubation time on the noise amplitude

We measured the logarithm of incubation time as a function of inverse squared noise amplitude. The results are presented at figure 2. One can see that logarithm of incubation time increases linearly with increasing inverse squared noise amplitude. Similar dependences were obtained for 5 supersaturations $\Delta c = c - c_{binodal}$: 0,1396; 0,1391; 0,1386; 0,1381; 0,1376.

3.2. Dependences of incubation time on supersaturation

We fixed some noise amplitude and varied the supersaturation within some interval which was not very wide since the decreasing supersaturation increases drastically the incubation time. At that, the computation becomes unreasonably long. Dependence of the logarithm of incubation time versus inverse squared supersaturation within the mentioned concentration interval is close to linear as shown at figure 3.

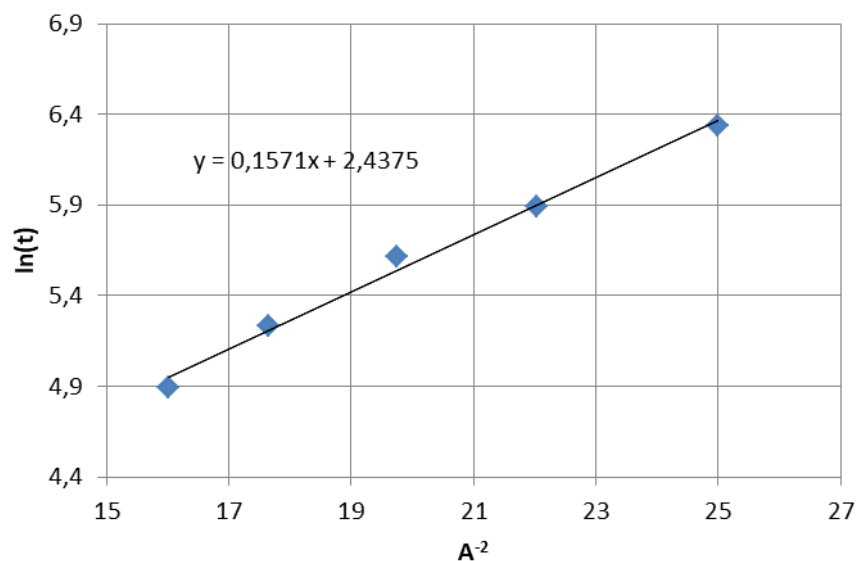


Fig. 2. Dependence of logarithm of incubation time on the inverse squared noise amplitude at reduced temperature $kT/V = 4$ and supersaturation $\Delta c = 0,1376$.

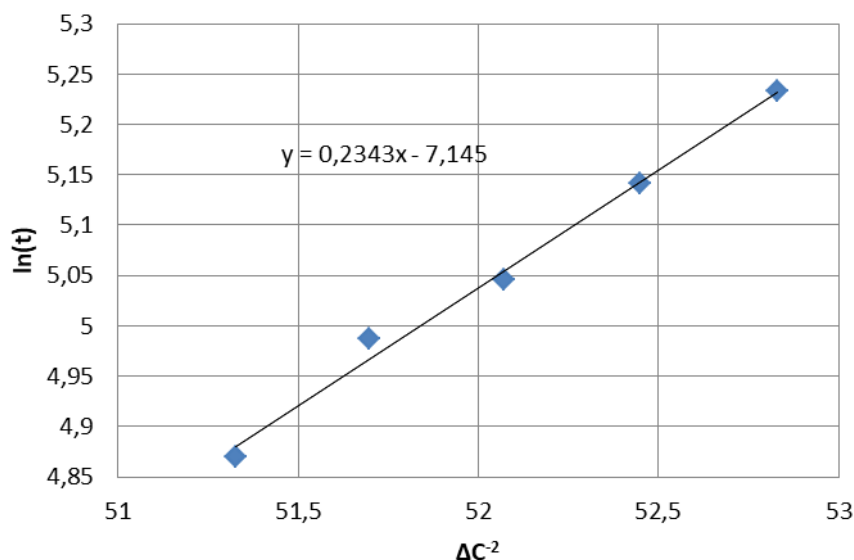


Fig. 3. Dependence of logarithm of incubation time on inverse squared supersaturation at fixed noise amplitude $A = 0,25$ and reduced temperature $kT/V = 4$.

4. Predictions of Classical Nucleation Theory

According to CNT, waiting time should be proportional to the exponent of reduced nucleation barrier. In its turn, the nucleation barrier is proportional to the cube of surface tension and inversely proportional to the squared driving force per atom of the nucleus. Using the theory of regular solid solution, it is easy to show that the interface surface tension along the coherent interface (100) is equal to:

$$\gamma = \frac{8}{a^2} (C^\beta - C^\alpha)^2 \left\{ \varphi_{AB} - \frac{\varphi_{AA} - \varphi_{BB}}{2} \right\} = \frac{8V_{mix}}{a^2} (C^\beta - C^\alpha)^2. \quad (6)$$

Here C^α and C^β are determined by the rule of a parallel tangent (not to confuse with the well-known rule of common tangent). The driving force can be reduced to the expression:

$$\Delta g = - \left(C_{binodal}^\beta - C_{binodal}^\alpha \right) \frac{\partial^2 g}{\partial C^2} \Delta C. \quad (7)$$

In CNT, the nucleus of the new phase appears as the result of heterophase nucleation with composition determined by a parallel tangent. Difference between two tangent points is practically independent on supersaturation. So, the surface tension of the nucleus is practically constant. On the other hand, the driving force is proportional to the supersaturation. Thus, the nucleation barrier in CNT should be inversely proportional to the squared supersaturation. This prediction coincides with SKMF results at figure 3.

5. Conclusions

Stochastic Kinetic Mean Field modeling on nucleation in supersaturated solution demonstrates the validity of Classical Nucleation Theory:

1. The nucleation process consists of two main steps: at first, the embryo of the new phase appears with almost optimal composition and then this embryo increases its size at almost constant composition.

2. The logarithm of nucleation time is inversely proportional to the squared supersaturation.

3. The logarithm of nucleation time is a linear function of the inverse squared noise amplitude.

Direct comparison with Monte Carlo simulation of nucleation will be presented elsewhere.

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**ЗАРОДКОУТВОРЕННЯ У МЕТАСТАБІЛЬНОМУ ТВЕРДОМУ РОЗЧИНІ –
ПОРІВНЯННЯ СТОХАСТИЧНОГО КІНЕТИЧНОГО СЕРЕДНЬОПОЛЬОВОГО
МЕТОДУ З КЛАСИЧНОЮ ТЕОРІЄЮ НУКЛЕАЦІЇ**

Анотація. Стадія зародкоутворення при фазових перетвореннях першого роду в сплавах має вирішальне значення для прогнозування механічних, електричних, магнітних властивостей багатофазних матеріалів. Протягом останніх десятиліть були розроблені нові експериментальні методи, що дозволяють безпосередньо спостерігати формування зародків під час твердофазних реакцій. Проте багато деталей зародкоутворення все ще залишаються нез'ясованими. Найбільш поширеними методами дослідження кінетики нуклеації є підхід Фоккера-Планка та моделювання методами Монте-Карло. Підхід Фоккера-Планка містить ряд невизначених параметрів і недоведених феноменологічних припущень. Монте-Карло методи є прямими і атомістичними, але рівень флуктуацій у них настільки високий, що досить важко ідентифікувати структури нових фаз у невеликих об'ємах.

У роботі представлено застосування Стохастичного кінетичного середньопольового методу (SKMF – Stochastic Kinetic Mean Field). Цей метод поєднує атомістичний підхід Жоржа Мартена з введенням шуму локальних атомних потоків. Підхід SKMF є нелінійним і тому може бути застосованим до опису ранніх стадій твердофазних реакцій у різкому градієнті концентрації. При такому підході ймовірність атомних обмінів пропорційна різниці експонент хімічних потенціалів, а не різниці хімічних потенціалів. Крім того, вводиться шум частот атомних стрибків замість шуму концентрацій. Моделювання нуклеації методом SKMF при розпаді перенасиченого бінарного твердого розчині свідчить про відповідність Класичній теорії нуклеації. Процес нуклеації складається з двох основних етапів: спочатку зародок нової фази з'являється з майже оптимальним складом, і збільшує свій розмір при майже постійному складі. Логарифм часу нуклеації обернено пропорційний квадрату перенасичення. Логарифм інкубаційного часу є лінійною функцією оберненого квадрату амплітуди шуму.

Ключові слова: SKMF метод, середнє поле, бінарний твердий розчин, розпад, зародкоутворення, інкубаційний час.

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